RESEARCH ARTICLE

M. Edmonds · R. A. Herd · B. Galle · C. M. Oppenheimer

Automated, high time-resolution measurements of SO₂ flux at Soufrière Hills Volcano, Montserrat

Received: 15 July 2002 / Accepted: 4 March 2003 / Published online: 17 May 2003 Springer-Verlag 2003

Abstract We report here the first results from an automated, telemetered UV scanning spectrometer system for monitoring SO_2 emission rates at Soufrière Hills Volcano, Montserrat. Two spectrometers receive light by way of a motor-driven stepping prism and telescope in order to make vertical scans of the volcanic plume. Spectral data from these spectrometers, situated 2,800 m apart and 4,500 m from the volcano, are relayed back to the observatory every 4–5 s via radio modems. A full scan of the plume is accomplished every 1–6 min by the (timesynchronised) spectrometers and a SO_2 emission rate is calculated using the $SO₂$ slant concentrations, scan angles and plume speeds estimated from the wind speed from a telemetered weather station near to the volcano. The plume's position and dimensions are calculated using the angular data from the two spectrometers. The plume height varies significantly diurnally and seasonally and is important in order to minimise the error on $SO₂$ emission rates. The new scanning system (Scanspec) provides SO_2 emission rates from 08:00 to 16:00 h local time every day. Preliminary results highlight a number of features of the $SO₂$ time series and plume dynamics and give our first indications of the errors and limits of detection of this system. $SO₂$ emission rates vary widely on all time scales (minutes, days, months). This new system has already provided the first real and consistent indication that $SO₂$ emission rates vary on a minutes to hours basis, which can be correlated with volcanic activity (for example, rockfall and pyroclastic flow activity). It is anticipated that this

Editorial responsibility: T.H. Druitt

M. Edmonds $(\mathbb{X}) \cdot R$. A. Herd Montserrat Volcano Observatory, Flemmings, Salem, Montserrat, West Indies e-mail: marie@mvo.ms Tel.: +1-664-4915647, Fax: +1-664-4912423

B. Galle

Chalmers University of Technology, Gothenburg, Sweden

C. M. Oppenheimer Department of Geography, Cambridge University, Downing Place, Cambridge, CB2 3EQ, UK

system at Soufrière Hills will yield information on shallow processes occurring on short time scales (periods of minutes to hours) as well as deep processes relating to magma supply rates, which will be associated with longer wavelength SO_2 signals of weeks to months.

Keywords Degassing · DOAS · Scanning spectrometers · Soufrière Hills, Montserrat · Sulphur dioxide emission rates

Introduction

Measurements of sulphur dioxide $(SO₂)$ emission rates have long been an integral part of monitoring networks in volcano observatories and the focus of research. However, nearly all the measurements to date have relied on labour-intensive field campaigns using the correlation spectrometer (COSPEC). This has limited observation periods to a few times per day at best. Where more sustained, high temporal resolution measurements have been attempted, correlations between SO_2 flux and volcanic activity have been observed (e.g. Kyle et al. 1994). At Soufrière Hills Volcano, Watson et al. (2000) identified a correspondence between $SO₂$ flux, seismicity and ground deformation by compositing the COSPEC data over several cycles of activity. Such data suggest that routine, high temporal resolution $SO₂$ flux monitoring could offer tremendous insights into degassing processes and, hence, hazard assessment. During 2001, a newly available, extremely compact ultraviolet spectrometer was used for the first time for volcanic SO_2 flux measurements, with encouraging results (Galle et al. 2003). The instrument clearly offered excellent scope for automation and real-time measurement and in this paper, a permanent installation for continuous, remote monitoring of $SO₂$ emission rates at a volcano is described. This scanspec system was installed in January 2002, with data telemetry from the scanning sites to the Montserrat Volcano Observatory (MVO) by way of radio modems. We describe here its operation for automated, real-time

fluxes from Soufrière Hills Volcano and present a selection of the preliminary dataset obtained and an estimate of the errors associated with it, highlighting aspects of high temporal resolution variability in observed fluxes and correlation with other monitored parameters and observed activity.

Soufriere Hills Volcano and prior $SO₂$ surveillance

Soufrière Hills Volcano, Montserrat, is an andesitic volcano in the Lesser Antilles island arc. It began to erupt in 1995, with a series of phreatic explosions in July 1995 (Young et al. 1998a; Robertson et al. 2000). Juvenile material was first erupted in November 1995 and formed a lava dome. Eruptive activity up to mid-1997 was comprised of dome building, with occasional small to moderate-sized $\left($ <10×10⁶ m³) collapses, generating pyroclastic flows. From August to October 1997, a series of vulcanian explosions occurred (Druitt et al. 2002). Thereafter, dome building continued to March 1998, interrupted by a sector collapse and lateral blast to the south-west in December 1997 (Sparks et al. 2002). A large collapse in July 1998 excavated the crater area and dome building recommenced in November 1999 (Norton et al. 2002). Since then, dome growth has continued and has been punctuated by two large collapses, involving 20 and 45 million cubic meters of dome material, on 20 March 2000 and 29 July 2001 respectively.

 $SO₂$ emission rate measurements provide insights into magma dynamics and chemistry on all time scales and are a valuable part of the risk assessment process at Soufrière Hills Volcano. Sulphur, exsolving (or partitioning) from magma at varying depths according to magma geochemistry, temperature and pressure (Scaillet et al. 1998), forms a fluid phase (with water, carbon dioxide and hydrogen chloride, among other minor species), which migrates upwards through the volcanic system (magma storage region and conduit) with time, carried in the magma as gas bubbles or migrating through a static body of magma through fracture and bubble networks. The arrival at the surface of this SO_2 -rich fluid phase depends strongly on magma chemistry and the permeability of the edifice (controlled by magma rise rates and sealing processes in the conduit and at the surface (Stix et al. 1993; Edmonds et al. 2003). The source of the $SO₂$ phase plays a large part in determining the signature of SO_2 emissions at the surface in relation to activity. $SO₂$ emissions from basaltic systems, such as Kilauea, Hawaii and Etna, Italy, for example, generally increase as large volumes of sulphur-rich magma approach the surface (e.g. Malinconico 1979; Chartier et al. 1987; Allard et. al. 1991; Caltabiano et al. 1994). Sulphur exsolves as the magma decompresses and eruptive events are preceded and accompanied by increased $SO₂$ emission rates, which rapidly decay once activity at the surface has ceased. Occasionally, the migration of batches of magma to shallow levels is not followed by eruptive activity, but these episodes can be recognised in the $SO₂$ time series.

At other more silicic volcanoes, such as Galeras, Colombia, for example, significant eruptive events have occurred with no prior change in $SO₂$ emission rates. Explosions at Galeras throughout 1991–1993 coincided with a long-term decrease in SO_2 emission rates as the magma supply system progressively degassed volatiles and the conduit gradually sealed (Stix et al. 1993; Fischer et al. 1997). The eruption of Mount St Helens, USA, in 1980, was preceded by a very low and steady SO_2 emission rate, which was later interpreted as a consequence of the low permeability of the caprock over the cryptodome, impeding gas release, which eventually failed, producing a lateral blast (Casadevall et al. 1981). $SO₂$ emissions during dome building at andesitic volcanoes such as Soufrière Hills, fluctuate, with high emission rates occurring when lava extrusion rates are high $(>3$ m^3 s⁻¹) and after large collapse events when the conduit is open, while low emission rates occur when dome growth stagnates or the conduit and dome system become sealed due to the precipitation of hydrothermal minerals (Edmonds et al. 2003).

 $SO₂$ emission rates have been measured at Soufrière Hills during much of the eruption, using a correlation spectrometer (COSPEC). The time series and its interpretation are discussed elsewhere (Oppenheimer et al. 1998, 2002; Young et al. 1998b, 2003; Watson et al. 2000; Edmonds et al. 2001, 2003). Whilst valuable information regarding long-term magma flux rates at depth and sealing processes at the surface have been gleaned from this dataset, the temporal resolution has been such that only general conclusions about processes happening on short time scales could be made. The source of the SO_2 -rich fluid phase at Soufrière Hills is thought to be the intrusion of a mafic magma, of basaltic-to-basaltic andesite composition, derived from mantle depths, which provided a thermal and chemical trigger for the eruption (Murphy et al. 1998; Couch et al. 2001). A clear SO_2 source signal from this mafic magma is distorted at the surface because of the rapid fluctuations in the permeability of the erupting andesite magma. The andesite undergoes vast changes in rheological properties in the upper conduit and dome due to degassing, cooling and crystallization (Sparks 1997; Cashman and Blundy 2000; Sparks et al. 2000). Fluctuating fluid pressure causes ephemeral changes in permeability as fracture and bubble networks open and close, which can cause rapidly changing emission rates of gas, on time scales of minutes to hours. The measurement of these short-term effects will be as revealing as the longer wavelength changes in SO2 emission rate caused by deep processes and are certainly as important for hazard assessment, yet are virtually unstudied. In particular, high temporal resolution monitoring of SO_2 emission rates on time scales of 1– 6 min, as presented here, can provide information on permeability and pressure changes in the conduit and dome that may be utilised in the routine monitoring of a volcano.

Phenomena such as hybrid earthquake swarms and banded tremor have occurred often during the dome

building stages of the Soufrière Hills eruption (White et al. 1998; Neuberg and O'Gorman 2002; Baptie et al. 2002) and have frequently been associated with increased pyroclastic flow activity and explosions. The $SO₂$ record may provide information on the source of these seismic events, helping constrain the feedback mechanisms between magma supply, degassing, microlite crystallisation, rheological properties and fluid pressure change and ultimately improve our understanding of how the magma and gas phases behave in the shallow volcanic environment. Previous studies of these phenomena have taken theoretical approaches (Denlinger and Hoblitt 1999; Melnik and Sparks 1999; Sparks et al. 2000). These models are corroborated, in part, by seismicity and ground deformation data and by observations. If the measurement of $SO₂$ emission rates can be improved to such an extent that the dataset has a comparable time resolution to the seismic and tilt data, then it is likely that it will help to refine these models still further.

Field installations of the new spectrometer system

An Ocean Optics S2000 spectrometer, coupled with an Ocean Optics ADC1000-USB A/D analogue to digital converter, is used at two field installations beneath the volcanic plume in Montserrat, one at Lovers Lane (LL) and one at Broderick's (BR; Fig. 1). The spectrometer utilises the Czerny–Turner optical design, which incorporates a high-sensitivity 2048-element linear CCD array detector. An optical fibre supplies light through a $50-\mu m$ slit and the light is then dispersed by way of a 2,400-line/ mm diffraction grating, over the array. The wavelength range is set at 280–420 nm. A similar type of spectrometer was first used on a volcano to measure SO_2 emission rate at Masaya, Nicaragua (Galle et al. 2003). The success of the technique prompted field trials in parallel with the COSPEC on Montserrat in August 2001. An excellent correlation between the two techniques was demonstrated (Galle et al. 2003) and plans were made for installing two such spectrometers, in fixed scanning mode, on Montserrat.

In order to scan through the plume, an optical system was built that sends light through a telescope into an optic fibre and a spectrometer. A stepper motor rotates a 90º reflecting prism, which reflects light into a telescope. Two lenses inside the telescope, 100 mm apart, focus the light into the optic fibre. The scanning system is illustrated in Fig. 2. The scanning systems are powered by solar panels and a 12-V battery. The spectrometer and the non-rotating part of the scanning system are housed in a weather-proof enclosure. FreeWave radio modems, operating at the (adjustable) baud rate of the spectrometer, transfer the data from each spectrometer back to the observatory and are also housed in the enclosure. The signal is repeated to get around topographical obstacles before reaching the observatory. Various electronic subsystems are required in order to modulate the voltage for the spectrometer to 5 V and to switch the power on in

Fig. 1 Map of southern Montserrat to show the locations of the two fixed scanning spectrometers at Lovers Lane and Brodericks and the weather station, providing wind (plume) speeds. The plume is shown in its most common azimuthal position

Fig. 2 The scanning and light collection devices used with both fixed scanning spectrometers at Soufrière Hills Volcano, Montserrat

the morning and off at night. The whole system consumes 8 W power during the daytime, dropping to less than 1 W when the spectrometers are powered down at night.

A UV spectrum from each of the spectrometers, encompassing the adjustable range 280–420 nm, is acquired at the observatory. The data acquisition system has been written and developed by MVO staff and is based on the serial command set provided by Ocean Optics for use with their spectrometers (Fig. 3). Data collection is programmed to begin at 08:00 and continues until 16:00 h, when the light levels are generally too low for spectral evaluation. The integration time (the time the CCD array is exposed to light) is adjusted automatically throughout the day by the software, in the range 200 to 1,000 ms, to account for changes in the intensity of sunlight throughout the day and to prevent saturation of the data stream. The integration time is linked to the angular velocity of the stepper motor such that, for any integration time, there are the same number of data points per scan (i.e. the prism rotates faster for faster measurement times). By averaging spectra on the spectrometer (before sending them back to the observatory), the level of signal to noise is improved (in proportion to the square root of the number of spectra averaged).

Fig. 3 Data acquisition software, developed by MVO. The display shows wavelength against intensity and the intensity of UV channel 315 nm against time for each spectrometer throughout the day for each spectrum

As the duration of a full revolution takes 80–400 s, the SO2 concentrations measured are not, in fact, a true instantaneous cross section of the plume. As the measurements in one scan are taking place, the plume is moving through this 2-D section at a speed equal to the wind speed at the plume height. Therefore, the time series does not sample mass fluxes every 80–400 s, rather the measurements represent an average mass flux throughout the measurement period. During periods of heightened activity on the volcano, the temporal resolution of measurements can be increased by reducing the bandwidths of the transmitted spectra. This decreases the data transfer time so that mass flux measurements can be made more frequently. Extensive error checking routines are incorporated into the software to reject spectra subject to telemetry or instrumental problems.

Plume azimuths are fairly constant in Montserrat. The plume drifts W-NW for 85–90 % of the year. The scanning sites are fixed in place and not easily moved for measurements on days when the plume is drifting in a different direction. Plans exist for another scanning spectrometer further to the NNW, which will improve the plume coverage. In the meantime, a smaller spectrometer (the Ocean Optics USB2000), is used for horizontal traverses beneath the plume, via helicopter,

when the plume is drifting out of range of the fixed scanning sites. This spectrometer is used with a laptop, via a USB link, in conjunction with a telescope mounted beneath the blades of the helicopter. The rotation of the helicopter blades does not affect the measurement. In this way, $SO₂$ emission rate data can be collected every day of the year, regardless of wind direction.

Spectral evaluation

When the system is 'awake', a UV spectrum is recorded every 4 or 5 s, from each spectrometer. These wavelength/intensity data are stored in separate data files. Another file is appended to for each scan, recording the time and the intensity of the 315 nm channel (which is absorbed by SO_2). Each spectrum is evaluated using the technique of differential optical absorption spectroscopy (DOAS), which is a way in which to eliminate atmospheric effects in the evaluation of the column concentrations for one gas species (Platt and Perner 1983; Platt 1994). The technique is based on the principle of isolating the narrow spectral features of the absorbent, by removing the broadband absorptions and attenuations due to Rayleigh molecular and Mie aerosol scattering. By

Fig. 4 The evaluated spectral output from the scanning system on Montserrat: SO_2 (ppm \times m) against time and SO_2 (ppm m) against scanning angle

measuring the 'differential' absorption, the method eliminates the need to identify and model absorption by air molecules. Absorptions can be split into those that vary slowly with wavelength (corresponding to atmospheric background absorption) and those that vary rapidly (due to the absorption of the molecular species of interest, $SO₂$ in this case). Normalisation of plume spectra with spectra containing no SO_2 , combined with the application of filtering techniques, eliminated the slow part of the absorption profile and isolates the $SO₂$ absorption feature, which can then be fitted to a library spectrum. The detailed procedure for the evaluation of the spectra collected by this system is as follows. A 'dark' spectrum (background electrical counts) is subtracted from the measured spectrum and then the measured spectrum is normalised by a 'blue sky' spectrum (away from the SO_2) plume). After normalisation, high and low pass filters are applied. The resulting logarithm of the $SO₂$ absorption feature, free from the sky spectrum and any non-volcanic SO_2 , is then fitted to a library reference spectrum of SO_2 (Vandaele et al. 1994), using a linear least squares procedure, whereby the measured spectrum is shifted and squeezed until the best fit is achieved and the slant column concentration is acquired by scaling the known concentration of the library spectrum. Preliminary evaluations were done using the software package DOASIS, developed by Heidelberg University, Germany. Subsequent evaluations have been carried out using customised software developed at MVO, incorporating the ability to evaluate two sets of spectral data, one from each spectrometer, using new 'no- SO_2 ' normalisation spectra for each scan, to account for changing atmospheric conditions throughout the day and then go on to 'pick' the plume from a zero baseline for each scan and calculate mass flux of $SO₂$. In this way, spectra can be evaluated and mass fluxes of $SO₂$ calculated continuously throughout the day. Figure 4 shows $SO₂$ ppm.m slant concentrations plotted against time and angle for several plume scans (the intensity of the 315 nm channel is also shown).

Mass flux calculation

The calculation of mass flux is based on the fixed COSPEC scanning procedure (Stoiber et al. 1983; Kyle et al. 1994). The calculation requires SO_2 slant columns versus angle (each measurement step is approximately 4.5º), plume speeds and directions (with time) and the distance to the plume vector (with time) throughout the period of measurement for each spectrometer. Plume speeds are notoriously difficult to obtain. At present we have a real-time, telemetered weather station, providing wind speed measurements at a sampling frequency of 4 Hz, which are then converted to minute averages, on top of St George's Hill (Fig. 1). The altitude of the station is around 350 m, which is approximately one-third of the height of the volcano. Plume heights are essential to constrain the measurements for the fixed scanning mode of $SO₂$ measurements. The inherent difficulty with this stationary scanning method for measuring concentrations is that a larger, higher plume will appear the same as a smaller, lower plume. An accurate estimate of plume height, therefore, is essential to minimise this error (Fig. 5). Two spectrometers collecting data simultaneous-

Fig. 5 For a horizontal traverse, plume height is not essential, as two plumes with the same mass at different heights (A and B) will produce equal SO_2 plume cross sections. For stationary scanning however, a bigger and higher plume (C) can look identical to a lower and smaller plume (D). Plume height is necessary to constrain these measurements

Fig. 6 Methodology for calculating plume height and position from the $SO₂$ concentration and angular data from the two fixed scanning spectrometers in Montserrat. Φ_1 and Φ_2 are calculated from Δ_1 and $\hat{\Delta_2}$, the scan angles for the peaks in SO₂ concentration measured during one revolution. Tan $\Phi_1=h/x_1$ and tan $\Phi_2=(h+50)/x_2$, where h is plume height. The height difference between the two spectrometers is 50 m (from GPS data) and $x_1+x_2=2,800$ m, the horizontal distance between them.

ly provide a method for calculating plume heights easily, using the maximum concentration vectors (Fig. 6).

The mass flux is currently generated as follows. The slant concentrations of $SO₂$ are calculated using the DOAS evaluation procedure described above, for both spectrometers. Usually one spectrometer is beneath the plume and one away from it and the two sets of data can be used to calculate the intersection of the two SO_2 concentration maxima vectors and hence the distance to and azimuth of the plume. The positions of the two spectrometers have been accurately determined using the differential Global Positioning System (GPS) and the angles of the full scan calibrated with theodolite measurements. The distance to the plume is used to generate a perpendicular plume width, using the 'picked' angular range. The integrated $SO₂$ concentration for the plume scan is multiplied by plume width, plume speed and a constant (to convert ppm units to mass units) and it is corrected for obliquity to the plume using the calculated azimuth. The result is the mass flux of SO_2 , in kg s⁻¹.

Errors

The sources of error identified on these measurements are listed below:

- 1. Spectrometer precision is less than 1% and the evaluation software will compensate for small shifts in pixel/wavelength drift, so this error is deemed negligible.
- 2. Error in the evaluation method. The evaluation procedure generates slant concentrations, not vertical column concentrations, which will lead to error on the

final flux calculations (we estimate this to be around 15%). This will be improved upon by incorporating air mass factors (dependent on wavelength and solar zenith angle) to calibrate the slant concentrations, computed with radiative transfer models.

- 3. Error in the DOAS method (smoothing, filtering, least squares fitting procedure). The error in this part of the procedure is primarily associated with the fitting routine and is small, estimated at 5%.
- 4. Errors on the calculation of plume width and azimuth (from scan angle and plume height). This has been minimised by calibrating the angular data using theodolite and accurately locating the spectrometers. An extra source of error lies in the fact that the plume exists at a range of heights and only the height of the $SO₂$ concentration maximum is used for the flux calculation. These errors total no more than 10%.
- 5. Errors in the estimation of plume speed in the mass flux calculation. The error on plume speed is estimated to be around -5% , $+30\%$ (i.e. the measured speed is most likely to be an underestimate of the true plume speed by around 30%, on the basis of comparisons with estimates of wind speed in the helicopter at plume height; Stoiber et al. 1983; Young et al. 1998b; Edmonds et al. 2003). Future improvements for estimating wind speed and direction will be based on incorporating radiosonde data into models of air-flow over topography (Wadge, personal communication). In the short term, a considerable improvement may be achieved by using a video camera to monitor the plume between fixed points in order to corroborate or scale the wind speed data from St George's Hill weather station.
- 6. Signal to noise changes throughout the day leading to operator errors in identifying or 'picking' the plume. The 'picking' of the plume will be subject to error when either the concentration of SO_2 in the plume is low or when the background noise is high (between 08:00–09:00 and 15:00–16:00 h, for example). This is likely to lead to an error ranging between 5 and 20%, with the larger error in the early morning and late afternoon.

The total errors will therefore be around -20 to $+36\%$ during most of the day and -28 to $+41\%$ during the early morning and late afternoon, when the signal to noise ratio is lower. The bias in the error is because there is a tendency to underestimate the plume speed rather than overestimate it. Errors will be reduced by using radiative transfer modelling, better estimates of plume speed and a greater number of scanning spectrometers. A proportion of the errors are systematic and likely to be constant for subsequent measurements, so the precision is considerably higher than the absolute accuracy of the measurements. Consequently, we estimate that changes in $SO₂$ emission rate throughout the day can be recognised with an error envelope of around -10 to $+30\%$. The limits of detection may be defined as three times the standard deviation (3σ) of the background noise, which corre-

Fig. 7 SO₂ emission rates from the spectrometers at Brodericks (grey line) and Lovers Lane (black line), calculated with accurate plume height data on 21 June 2002. Error bars are –20 to 36% for most of the measurements (see text for discussion of errors)

sponds to the minimum recognisable departure from the baseline and consequently the limit of detection is in the range 5–10 ppm.m, a slant concentration–path length product. The limit of detection will be higher in the early morning and late afternoon and also during conditions of rain or opaque ash clouds, which leads to a lower intensity of light entering the spectrometer and the signal to noise ratio is decreased. When combined with a low wind speed (3 m/s) and an average plume configuration, this limit of detection translates to a minimum detectable mass flux of $SO₂$ of 0.3 to 0.7 kg/s (25–60 t/day), with the higher detection limit corresponding to early morning and late afternoon.

Preliminary results and interpretation

The scanning spectrometers have been fully operational since March 2001. The preliminary results from this first continuous monitoring highlight a number of interesting features of SO_2 emission at Soufrière Hills Volcano.

Figure 7 shows a 4-h period of flux data, calculated using both spectrometers. Spectrometer data from both sites were used to calculate the distance to the plume vector and width perpendicular to this vector (for each scan) and this plume configuration was used to calculate an emission rate for both sets of spectrometer data. $SO₂$ emission rates vary throughout the course of one day, sometimes increasing or decreasing steadily, at other times showing an oscillation of varying periods, typically 60–180 min. Figure 8 shows some examples. RSAM

Fig. 8 $SO₂$ emission rates calculated using data from both spectrometers, plotted against time for 24 April, 23 April, 25 March and 26 April 2002 (grey points and trend line). Also shown is a 20-min period moving average trend line (black line) of the relative seismic amplitude measurement (RSAM) from the seismometer at Windy Hill, Montserrat

Fig. 9 Mean SO_2 emission rate throughout June 2002 (grey squares with a trend line), shown with –20 to 36% error bars. The black line is a 20-point moving average of the 30-min RSAM from the Windy Hill seismometer, for comparison with $SO₂$ data

(relative seismic amplitude measurement) is also shown for comparison to the SO_2 emission rate data. A correlation between SO_2 emission rate and seismicity is evident for parts of the time series. These seismic data represent the raw seismic signal and consist mainly of surficial events such as rockfalls, with the bigger signals representing pyroclastic flows. It appears that $SO₂$ emissions broadly follow trends in rockfall and pyroclastic flow activity. This is to be expected, as the failure and fragmentation of large blocks from the dome will lead to the release of SO_2 trapped in pore spaces and closed fracture networks. The $SO₂$ data here are representative of gas flow through a large (300 m high and 500–600 m wide) dome; therefore, we would expect the signal to be modulated to a great degree by the variably permeable andesite mass. Ephemeral increases in permeability occur when blocks break off from the dome and when fluid pressure build-up causes fracture networks to open and close periodically, thus creating a feedback between pressure build-up, permeability change and gas release. This is an aspect of shallow volcanic behaviour in which our new SO_2 monitoring system will become increasing valuable, allowing the correlation of these patterns in gas release with other datasets.

Plume heights and azimuths have proved to be highly variable, especially in the early mornings and late afternoon and so have to be carefully evaluated in the flux calculation method. The behaviour of smoke stack plumes under varying atmospheric conditions has been studied extensively (e.g. Beychok 1995) and may be applied to volcanic plumes. The plume from Soufrière Hills Volcano often fumigates (flows close to the ground), lofts (rises nearly vertically up from the vent), puffs and bifurcates (breaks up into two or more separate plumes, with different azimuths, caused by separate 'puffs' rising in the centre and descending on the sides). These configurations are largely determined by the atmospheric conditions and will vary throughout a day and from rainy season to dry season. The measurement of plume height

and azimuth for the calculation of SO_2 emission rates from scanning data, therefore, is essential.

Longer wavelength $SO₂$ emission rate signals have been seen in the COSPEC data from Montserrat and are related to deep magma supply rates. The new SO_2 monitoring system presented here is expected to highlight these features more accurately over time, after extracting the higher frequency, near-surface influences. Figure 9 shows the daily mean $SO₂$ emission rates for June 2002, shown with the smoothed 30-min RSAM. The volcano was relatively inactive over this period, which is reflected in the low mean $SO₂$ emission rate. Peaks and troughs in $SO₂$ emission rate over timescales of weeks to months may indicate changes in magma supply to the upper conduit.

Conclusions

The availability of cheap, portable and accurate UV spectrometers, utilising new CCD array technology, will change the way in which $SO₂$ (and other gas) emissions are monitored at volcanoes. It is expected that these advances will increase the value of gas monitoring to a point at which the study of $SO₂$ emissions will yield fine insights into magma flux rate from depth and near-surface processes of permeability and fluid pressure changes. Preliminary results obtained from automated, high temporal resolution SO_2 monitoring illustrate the great potential of such an approach for day-to-day assessment of eruptive hazards. This is a dramatic improvement over previous COSPEC-based surveillance of this volcano and will lead to new perspectives for SO_2 monitoring at other volcanoes, worldwide.

Acknowledgements This paper is published with the permission of the director of the British Geological Survey, UK, and Peter Dunkley is acknowledged for his review. Patrick Allard is thanked for his excellent review, which improved the paper considerably. MVO and BGS are acknowledged for permission to publish these data. C.M.O. gratefully acknowledges support from the EC Framework 5 project 'MULTIMO'. The use of trade names in this paper does not imply endorsement by MVO. Rob Waterbury, from Ocean Optics, is acknowledged for his invaluable advice.

References

- Allard P, Carbonnelle J, Dajlevic D, Le Bronec J, Morel P, Maurenas JM, Robe MC, Faivre-Pierret R, Sabroux JC, Zettwoog P (1991) Eruptive and diffuse emissions of carbon dioxide from Etna volcano. Nature 351:387–391
- Baptie B, Luckett R, Neuberg J (2002) Variability of seismic tremor episodes at Soufrière Hills Volcano, Montserrat. In: Druitt TH, Kokelaar BP (eds) The eruption of the Soufrière Hills Volcano, Montserrat, from 1995 to 1999. Geological Society London, Memoir, pp 611–620
- Beychok MR (1995) Fundamentals of stack gas dispersion, 3rd edn. Milton R Beychok, Irvine, California
- Caltabiano T, Romano R, Budetta G (1994) SO₂ flux measurements at Mount Etna, Italy. J Geophys Res 99:12809–12819
- Casadevall TJ, Johnston DA, Harris DM, Rose WI, Malinconcio LL, Stoiber RE, Bornhorst TJ, Williams SN, Woodruff, Laurel,

Thompson JM (1981) $SO₂$ emission rates at Mount St. Helens from March 29 through December, 1980. In: Lipman PW, Mullineaux DL (eds) The 1980 eruptions of Mount St. Helens, Washington. US Geol Surv Prof Pap 1250:193–200

- Cashman K, Blundy J (2000) Degassing and crystallisation of ascending andesite and dacite. Phil Trans R Soc Lond A 358:50–54
- Chartier TA, Rose WI, Stokes JB (1987) Detailed record of $SO₂$ emissions from Pu`u` \hat{O} between episodes 33 and 34 of the 1983–86 east rift zone eruption of Kilauea, Hawaii. Bull Volcanol 50:215–228
- Couch S, Sparks RSJ, Carroll MR (2001) Mineral disequilibrium in lavas explained by convective self-mixing in open magma chambers. Nature 411:1037–1039
- Denlinger RP, Hoblitt RP (1999) Cyclic eruptive behaviour of silicic volcanoes. Geology 27:459–462
- Druitt TH, Young SR, Baptie B, Bonadonna C, Calder ES, Clarke AB, Cole PD, Harford CL, Herd RA, Luckett R, Ryan G, Voight B (2002) Episodes of Vulcanian explosive activity with fountain collapse at Soufrière Hills Volcano, Montserrat. In: Druitt TH, Kokelaar BP (eds) The eruption of the Soufriere Hills Volcano from 1995 to 1999, Montserrat, Antilles. Memoir of the Geological Society of London, pp 281–306
- Edmonds M, Pyle DM, Oppenheimer CM (2001) A model for degassing at Soufrière Hills Volcano, Montserrat, West Indies based on geochemical data. Earth Planet Sci Lett 186:159–173
- Edmonds M, Oppenheimer C, Pyle DM, Herd RA, Thompson G (2003) SO₂ emissions from Soufrière Hills Volcano and their relationship to conduit permeability, hydrothermal interaction and degassing regime. J Volcanol Geotherm Res 124 (1-2): 23- 43. DOI:10.1016/S0377-0273(03)00041-6
- Fischer TP, Arehart GB, Sturchio NC, Williams SN (1997) The relationship between fumarole gas composition and eruptive activity at Galeras Volcano, Colombia. Geology 24:531–534
- Galle B, Oppenheimer CM, Geyer A, McGonigle A, Edmonds M (2003) A mini-DOAS spectrometer applied in remote sensing of volcanic SO2 emissions. J Volcanol Geotherm Res 119(1– 4):241–254
- Kyle PR, Sybeldon LM, McIntosh WC, Meeker K, Symonds RB (1994) Sulfur dioxide emission rates from Mount Erebus, Antarctica. In: Kyle PR (ed) Volcanological and environmental studies of Mount Erebus, Antarctica. Antarctic Res Ser 66:69– 82
- Malinconico LL (1979) Fluctuations in $SO₂$ emission during recent eruptions of Etna. Nature 278:43–45
- Melnik O, Sparks RSJ (1999) Non-linear dynamics of lava dome extrusion. Nature 402:37–41
- Murphy MD, Sparks RSJ, Barclay J, Carroll MR, Lejeune AM, Brewer TS, Macdonald R, Black S, Young S (1998) The role of magma mixing in triggering the current eruption at the Soufrière Hills volcano, Montserrat, West Indies. Geophys Res Lett 25(18):3433–3436
- Neuberg J, O'Gorman C (2002) A model of the seismic wavefield in gas-charged magma: application to Soufrière Hills Volcano, Montserrat. In: Druitt TH, Kokelaar BP (eds) The eruption of the Soufrière Hills Volcano, Montserrat, from 1995 to 1999. Geological Society London, Memoir pp 603–610
- Norton GE, Herd RA, Watts R, Aspinall WP, Young SR, Bonadonna C, Baptie B, Edmonds M, Harford C, Jolly AD, Loughlin SC, Luckett R, Mattioli G, Sparks RSJ (2002) Pyroclastic flow and explosive activity of the lava dome of Soufrière Hills Volcano, Montserrat, during a period of no magma extrusion (March 1998 to November 1999). In: Druitt TH, Kokelaar BP (eds) The eruption of the Soufrière Hills Volcano, Montserrat, from 1995 to 1999. Geological Society London, Memoir pp 467–482
- Oppenheimer CM, Francis PW, Stix J (1998) Depletion rates of SO2 in tropospheric plumes. Geophys Res Lett 25:2671–2674
- Oppenheimer CM, Edmonds M, Francis P, Burton M (2002) Variation in HCl:SO₂ gas ratios at Soufrière Hills Volcano observed by Fourier Transform Spectroscopy, 1996–1999. In: Druitt TH, Kokelaar BP (eds) The eruption of the Soufriere Hills Volcano from 1995 to 1999, Montserrat, Antilles. Geological Society London, Memoir pp 621–639
- Platt U (1994) Differential optical absorption spectroscopy (DOAS). In: Sigrist MW (ed) Air monitoring by spectroscopic techniques, vol 127. Wiley, Chichester, pp 27–84
- Platt U, Perner D (1983) Measurements of atmospheric trace gases by long path differential UV/visible absorption spectroscopy. Springer, Berlin Heidelberg New York, Ser Opt Sci 39:95–105
- Robertson REA, Aspinall WP, Herd RA, Norton GE, Sparks RSJ, Young SR (2000) The 1995–98 eruption of the Soufrière Hills volcano, Montserrat, WI. Phil Trans R Soc Lond 358(1770):1619–1638
- Scaillet B, Clemente B, Evans BW, Pichavant M (1998) Redox control of sulphur degassing in silicic magmas. J Geophys Res 103:23937–23949
- Sparks RSJ (1997) Causes and consequences of pressurisation in lava dome eruptions. Earth Planet Sci Lett 150:177–189
- Sparks RSJ, Murphy MD, Lejeune AM, Watts RB, Barclay J, Young SR (2000) Control on the emplacement of the andesite lava dome of the Soufrière Hills volcano by degassing-induced crystallisation. Terra Nova 12:14–20
- Sparks RSJ, Barclay J, Calder ES, Herd RA, Luckett R, Norton GE, Ritchie LJ, Voight B, Young SR, Woods AW (2002) Generation of a debris avalanche and violent pyroclastic density currents on 26 December (Boxing Day) 1997 at Soufrière Hills Volcano, Montserrat. In: Druitt TH, Kokelaar BP (eds) The eruption of the Soufrière Hills Volcano, Montserrat, from 1995 to 1999. Geological Society London, Memoir pp 409–434
- Stix J, Zapata GJA, Calvache VM, Cortes JGP, Fischer TP, Gomez MD, Narvaez ML, Ordonez VM, Ortega EA, Torres CR, Williams SN (1993) A model of degassing at Galeras Volcano, Colombia, 1988–1993. Geology 21:963–967
- Stoiber RE, Malinconico LL, Williams SN (1983) Use of the correlation spectrometer at volcanoes. In: Tazieff H, Sabroux JC (eds) Forecasting volcanic eruptions. Elsevier, Amsterdam, pp 425–444
- Vandaele AC, Simon PC, Guilmot JM, Carleer M, Colin R (1994) $SO₂$ absorption cross section measurements in the UV using a Fourier transform spectrometer. J Geophys Res 99:25599– 25605
- Watson IM, Oppenheimer C, Voight B, Francis PW, Clarke A, Stix J, Miller AD, Pyle DM, Burton MR, Young SR, Norton G, Loughlin S, Darroux B (2000) The relationship between degassing and ground deformation at Soufrière Hills volcano, Montserrat. J Volcanol Geotherm Res 98:117–126
- White RA, Miller AD, Lynch LL, Power J (1998) Observations of hybrid seismic events at Soufriere Hills Volcano, Montserrat: July 1995 to September 1996. Geophys Res Lett 25(19):3657– 3660
- Young SR, Sparks RSJ, Aspinall WP, Lynch LL, Miller AD, Robertson REA, Shepherd JB (1998a) Overview of the eruption of Soufrière Hills Volcano, Montserrat, July 18, 1995, to December 1997. Geophys Res Lett 25(18):3389–3392
- Young SR, Francis PW, Barclay J, Casadevall T, Gardner CA, Darroux B, Davies MA, Delmelle P, Norton GE, Maciejewski AAM, Oppenheimer CM, Stix J, Watson IM (1998b) Monitoring SO_2 emission at the Soufrière Hills Volcano: implications for changes in eruptive conditions. Geophys Res Lett 25(19):3681–3684
- Young SR, Voight B, Duffell HJ, Baptie BP, Jolly AD, Syers RC, Thompson G (2003) Magma extrusion dynamics revealed by high-frequency gas monitoring at Soufrière Hills Volcano, Montserrat. J Geol Soc Lond Spec Publ (in press)